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13. ABSTRACT (Maximum 200 words) A home made semi-automatic feeding plasma arc reactor was used for the mass production of fullereness. Large, high quality single crystals of C_{60} up to 3mm in length have been grown by both the open-end and sealed vapor transport techniques. Millimeter-sized superconducting single crystals of K_3C_{60} were successfully produced by a sophisticated vacuum doping method. Magnetic and superconducting properties were obtained. Information on the field and temperature dependences of the critical current density J_c and the H_{c1} have been reported. New compounds of Yb_xC_{60} with $x=1-6$ were synthesized using a liquid ammonia route at low T and in an inert atmosphere. It turns out that these compounds are amorphous and have a spin glass behavior at a temperature $T < 15K$. Finally, new superconductors with T_c of 134K were found in the system of $Tl_{1-x}Hg_xBa_2Ca_2Cu_3O_{8+y}$. This invention has great potential for future applications.				
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Final Technical Report (Oct., 1992-Sept., 1995)

(1) Research objectives:

- a) Growth of large high quality of single crystal of C_{60} and superconducting A_3C_{60} , where A represents alkali metal.
- b) Extensive physical measurements on the normal-state and superconducting properties of C_{60} and A_3C_{60} .
- c) Search for new, air stable magnetic and superconducting fullerides.
- d) Synthesis and study of the Hg-1223 high temperature superconductors.

(2) Status of the research efforts:

- a) A modified plasma arc reactor for the mass production of fullerenes was designed and manufactured. The reactor employs mechanisms for continuous graphite-rod feeding and in-situ slag removal. Production of 10 gram of soot per hour is readily achieved utilizing this reactor. Fullerene yields up to 20% are attained routinely.
- b) A new vapor transport technique to grow single crystals of C_{60} was developed. With this technique, high quality C_{60} crystals with linear dimension of 1-3 mm was obtained.

- c) Thin films of C_{60} with thickness up to several microns were fabricated by direct vacuum evaporation.
- d) A simplified technique for synthesizing high quality millimeter-sized single crystals of K_3C_{60} and Rb_3C_{60} was developed. The preparation method consists a two-step process where pure C_{60} crystals are first grown and later doped with Potassium or Rubidium vapor. A specialized quartz tube was designed to allow superconducting phase purity to be monitored via low field shielding magnetization measurements.

Magnetic properties of the K_3C_{60} and Rb_3C_{60} crystals have been measured by using a SQUID magnetometer. Data on the field and temperature dependences of the critical current density J_c and H_{c1} are obtained, which is quite different from the results obtained from polycrystalline samples. Comparisons between K_3C_{60} and Rb_3C_{60} indicate that defects may be more prevalent in the latter compound due to more sensitive synthesis requirements. Also, time relaxations of the magnetizations of the K_3C_{60} and Rb_3C_{60} have been conducted to study the dynamics of the flux motions.

- e) The magnetic and structural properties of C_{60} interstitially doped with the rare earth ytterbium have been studied. Samples of Yb_xC_{60} with nominal stoichiometries ranging from $x=1-6$ were synthesized in cooperation with the Department of Chemistry. A low temperature liquid ammonia route was used to intercalate the rare earth ions into the fullerene

lattice. Synthesis was carried out under an inert atmosphere to reduce potential contamination. The product had an amorphous structure that decomposes into C_{60} and Yb_2O_3 if heated in air. Magnetic measurements showed that the Yb_xC_{60} exhibits Curie-Weiss paramagnetism for $T > 15K$ with ytterbium ions in a 3+ oxidation state. At low temperature ($T < 15K$), a spin glass state was found in these compounds.

- f). A new system of superconductors of $Tl_{1-x}Hg_xBa_2Ca_2Cu_3O_{8+\delta}$ (TlHg-1223) with $0.1 \leq x \leq 1.0$ and $T_c > 130K$ was developed.

Polycrystalline samples consisting of a near pure superconducting phase of $T_c = 134K$ were fabricated over a broad composition range. Powder x-ray diffraction showed a continuous solubility exists in this system. Millimeter size single crystals of TlHg-1223 with T_c of 134K were also produced by a liquid phase promoted solid state reaction technique. Based on this progress, researchers now can use near phase pure polycrystal specimens or single crystals to study the various properties of these record high T_c superconductors.

(3) Publication list

- 1) J.Z. Liu, J.W. Dykes, M.D. Lan, P. Klavins, R.N. Shelton, and M.M. Olmstead. " Vapor transport growth of C_{60} crystals". Appl. Phys. Lett. 62, 531(1993).
- 2) M. Bennahmias, A.F. Bello, D. Back, H.B. Radousky, T.J. Goodwin, P. Klavins, and R.N. Shelton. " Magnetic

- properties of polycrystalline $R_{1.5}Ce_{0.5}Sr_2Cu_2NbO_{10}$
($R=Eu, Nd$ and Sm) high- T_c superconducting ceramics" Phys.
Rev. B48, 6525(1993).
- 3) P.E. Anderson, T.T. Anderson, P.L. Dyer, J.W. Dykes, S.H. Irons, C.A. Smith, R.D. Kylin, P. Klavins, J.Z. Liu, and R.N. Shelton. " Optimization of Fullerene yields in a plasma Arc reactor". Proc. of the Recent Advances in the Chemistry and Physics of Fullerenes and Related Materials, P.40 (1994), The Electrochemical Society, Proceedings Volume 94-24, 40(1994).
 - 4) J.W. Dykes, P. Klavins, M.D. Lan, J.Z. Liu, and R.N. Shelton. " Production of Single crystal K_3C_{60} ". J. of Superconductivity, 7:635(1994).
 - 5) S.H. Irons, J.Z. Liu, P. Klavins, M.D. Lan, R.N. Shelton, K. Song, S.M. Kauzlarich. " Synthesis and Properties of Ytterbium doped C_{60} ". Proc. Mat. Res. Soc., 349,307(1994).
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 - 7) M.D. Lan, J.Z. Liu, Y.X. Jia, Lu Hang, and R.N. Shelton. "Normal-State Hall effect in $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ Single Crystals". Phys. Rev. B49, 580(1994).
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- Studied by Torque Magnetometry".** Phys. Rev. B49, 1342(1994).
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 - 10) H. Buan, B. Zhou, S.W. Pierson, C.C. Huang, O.T. Valls, J.Z. Liu, and R.N. Shelton. "Analysis of the Specific Heat of High- T_c Superconductors in Magnetic Fields". Physica B194-196, 1491(1994).
 - 11) J. Buan, Branko P. Stojkovic, N.E. Israeloff, A.M. Goldman, C.C. Huang, Oriol T. Valls, J.Z. Liu and Robert Shelton. "Transverse Magnetization Study of the Pairing State of the High- T_c Superconductor $LuBa_2Cu_3O_{7-\delta}$ ". Phys. Rev. Lett. 72, 2632(1994).
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 - 13) I.C. Chang, M.D. Lan, T.J. Goodwin, J.Z. Liu, and R.N. Shelton. "Magnetic and Transport Properties of $HgBa_2Cu_4-\delta$ ". Physica C223,207(1994).
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- Single Crystal". Phys. Rev. B50, 7092(1994).
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 "Magnetic-Relaxation Study in Melt-Textured $\text{ErBa}_2\text{Cu}_3\text{O}_{7-\delta}$ Ceramic Material". Applied Physics A59, 169(1994).
- 17). J.W. Dykes, W.D. Mosley, P.A. Sterne, J.Z. Liu, R.N. Shelton, R.H. Howell, "Investigation of the electronic distribution in the octahedral sites of solid C_{60} through the 260K orientational phase transition" Chemical Physics Letters, 232, 22(1995)
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(4) List of professional Personnel

J.Z. Liu	Research Physicist II, PI
R.N. Shelton	Professor VIII
P. Klavins	Specialist II
T.T. Anderson	Engineer
P.L. Dyer	Mechanics
J.W. Dykes	Graduate student
S.H. Irons	Graduate student
P.E. Anderson	Graduate student
I.C. Chang	Graduate student
C.A. Smith	Undergraduate student

(5) Interactions(Coupling activities)

a) Papers presented at meetings, conferences, seminars, etc.

1. J.Z. Liu, **Vapor transport growth of C_{60} crystals.** MRS Fall Meeting, Boston, 1993.
2. J.W. Dykes, **Production of Single crystal K_3C_{60} .** presented at "Physics and Chemistry of Molecular and Oxide Superconductors"-a satellite conference to LT20, Eugene, Oregon, July 27-31, 1993.
3. J.Z. Liu, **Growth and Characterization of Superconducting Single Crystals.** A invited seminar tour at the following Universities(February-March, 1993):
National Tsing Hua University, Taiwan, ROC
National Taiwan University, Taiwan, ROC
National Chung Cheng University, Taiwan, ROC

National Chung Shan University, Taiwan, ROC

Industry Technology Research Institute, Taiwan, ROC

Hong Kong University, Hong Kong

Hong Kong University of Science and Technology, Hong Kong

Nanjing University, Nanjing, PRC

4. S.H. Irons et al. "Synthesis and properties of Ytterbium doped C_{60} ". MRS Spring Meeting, San Francisco, 1994.
5. P. Klavins et al. " Characterization of the Arc-Process for Fullerene Production". presented at MRS Fall meeting, Boston, 1993.
6. P. Klavins et al. " Optimization of Fullerene Yields in a Plasma ARC Reactor". Invited talk at 1994 Spring Electrochemical Society Meeting, San Francisco.
7. S.H. Iron et al. " Synthesis of K_3C_{60} from Single Crystal C_{60} and Determination of its Superconducting Parameters", presented in APS 95 March meeting, San Jose, CA.

b) Consultive and advisory functions to other parties.

None

(6) New discoveries.

New superconductors with T_c above 130K were discovered in the system of $Tl_{1-x}Hg_xBa_2Cu_3O_{8+\delta}$. This invention was disclosed to the University of California Technical Transfer Office. (University of California patent case No.94-061-1).

(7) Other statements. None